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ELECTROMAGNETIC SCALE MODELS USING EMULSIONS

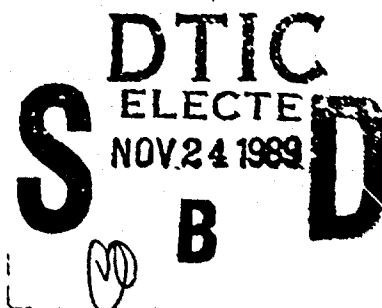
Georgia Institute of Technology

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This emulsion matches the electrical constitutive parameters of the clay, including the dispersion in the conductivity, over a ten-to-one frequency range.

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INTRODUCTION

The research on this contract involved the study of emulsions as materials for use in electromagnetic scale models. A major portion of the effort was a systematic experimental investigation that involved the formulation and testing of several hundred emulsions. The goal of this investigation was to develop a series of stable emulsions with predictable electrical constitutive parameters (relative permittivity and conductivity) that covered a wide range of values. This goal was accomplished in the manner described in the accompanying paper entitled, "The Use of Emulsions to Represent Dielectric Materials in Electromagnetic Scale Models". This paper was submitted for journal publication, and an abbreviated version of this paper was presented at the IEEE International Symposium on Antennas and Propagation, Syracuse, NY, June 1988.



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THE USE OF EMULSIONS TO REPRESENT DIELECTRIC
MATERIALS IN ELECTROMAGNETIC SCALE MODELS

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ABSTRACT

The electrical constitutive parameters of materials in a scale model and the corresponding full-sized system must satisfy certain relationships. Thus, it is desirable to have a series of materials with a range of electrical parameters or mixtures of materials with adjustable electrical parameters for use in scale models. In this paper, simple emulsions are examined as materials with adjustable electrical constitutive parameters. These emulsions are mixtures of oil, saline solution, and a suitable stabilizing agent (emulsifier). Since the relative permittivities of oil and water are around two and eighty, respectively, a large range of permittivity can be obtained for the emulsions. The conductivity of the emulsions can be adjusted by changing the normality of the saline solution. A series of oil-in-water emulsions (oil droplets in water), suitable for use in scale models, is developed; this includes the selection of an appropriate emulsifier. The electrical constitutive parameters of these emulsions are adjustable over wide ranges, and are predictable from a simple formula. As an example, an emulsion that is a scale model for red-clay earth is described. This emulsion matches the electrical constitutive parameters of the clay, including the dispersion in the conductivity, over a ten-to-one frequency range.

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I. INTRODUCTION

Electromagnetic scale models are of value in the design and testing of electromagnetic systems that are on a scale too large or too small for routine laboratory investigation [1,2]. For example, the performance of antennas over or buried in the earth can be evaluated in the laboratory using reduced size scale models operated at frequencies higher than those used for the actual antennas. For this application, a material is needed that will model the electrical properties of the earth.

A common form for the scale model is one in which the physical dimensions (\vec{r}) are scaled by the factor k_L and the frequency (ω) is scaled by the factor $k_\omega = 1/k_L$:

$$\vec{r}_m = k_L \vec{r}_f \quad (1a)$$

$$\omega_m = k_\omega \omega_f = (1/k_L) \omega_f, \quad (1b)$$

where the subscripts f and m refer to the full-sized system and the model, respectively [1]. The electrical constitutive parameters for every material in the system must also be scaled. The scale factors are, for the permittivity $k_\epsilon = 1$

$$\epsilon_m(\omega_m) = k_\epsilon \epsilon_f(\omega_f) = \epsilon_f(\omega_f), \quad (1c)$$

for the permeability $k_\mu = 1$

$$\mu_m(\omega_m) = k_\mu \mu_f(\omega_f) = \mu_f(\omega_f), \quad (1d)$$

and for the conductivity $k_\sigma = 1/k_L$

$$\sigma_m(\omega_m) = k_\sigma \sigma_f(\omega_f) = (1/k_L) \sigma_f(\omega_f). \quad (1e)$$

From (1c) the permittivities of the materials in the full-sized system and the model must be the same. Note however, that the right-hand side of (1c) is evaluated at the frequency ω_f while the left-hand side is evaluated at the frequency ω_m . Consequently, a material in the full-sized system can only be used to represent itself in the model when the permittivity of the material does not have significant dispersion over the frequency range which includes ω_f and ω_m . The condition (1d) for the permeabilities is easily met when all materials in the full-sized system and model are nonmagnetic $\mu = \mu_0$. The conductivities (1e) in the full-sized system must be scaled by the factor $1/k_f$ in the model.

Clearly it is desirable to have a series of materials with a range of electrical properties or mixtures of materials with adjustable electrical properties for use in scale models. Examples of the latter are saline solutions at frequencies below the microwave range; the solutions have a nearly constant permittivity and a conductivity that is adjustable by varying the salt concentration. Mixtures of materials that have both an adjustable permittivity and an adjustable conductivity are much more difficult to formulate. In addition to having the desired electrical properties, materials used in scale models should be nontoxic, nonflammable, noncorrosive, and relatively inexpensive.

In this paper, emulsions, which are mixtures of oil and water, are examined as materials for use in scale models. The relative permittivity of oil is around two, while that of water is around eighty. Thus, a wide range of relative permittivity can be obtained by mixing these components. The conductivity of the emulsion can be adjusted by changing the concentration of an electrolyte, such as sodium chloride, in the aqueous phase.

II. BASIC PROPERTIES OF EMULSIONS

"An emulsion is a heterogeneous system, consisting of at least one immiscible liquid intimately dispersed in another in the form of droplets, whose diameters, in general, exceed 0.1μ . Such systems possess a minimal stability, which may be accentuated by such additives as surface-active agents, finely-divided solids, etc." [3].

The prototypical emulsion is one where the two phases are oil and water, and the discussion presented here will be limited to this case. When the disperse phase (droplets) is oil and the continuous phase (matrix) is water, the emulsion is referred to as an oil-in-water emulsion (O/W). Conversely, when the disperse phase is water and the continuous phase is oil, the emulsion is referred to as a water-in-oil emulsion (W/O). Two familiar emulsions are milk and butter; milk being an O/W emulsion, while butter is a W/O emulsion. Other common products that are emulsions include: cosmetic creams and lotions, pharmaceuticals, polishes, paints, asphalt, and numerous food preparations.

Ordinary experience suggests that oil and water do not readily form a stable mixture. The formation of an emulsion usually requires the addition of an emulsifier or surface-active agent and substantial agitation [3-5]. The molecules of the emulsifier are absorbed at the interface between the two immiscible liquids (oil/water interface). The emulsifier lowers the interfacial surface tension between the liquids. While this greatly aids in the formation of an emulsion, it may not be the only significant effect of the emulsifier [3].

Generally, the molecules of the emulsifier have a hydrophilic (water-loving) group which is oriented toward the water side of the interface, and a lipophilic (oil-loving) group which is oriented toward the oil side of the interface. The emulsifier may be anionic, cationic, or nonionic. For anionic (cationic) emulsifiers, anions (cations) are absorbed at the oil/water interface and cations (anions) are released into the aqueous phase, while for nonionic emulsifiers, few free ions are released into the aqueous phase.

The effectiveness of an emulsifier is greatly dependent on the ingredients of the emulsion, and an empirical procedure is generally used to select the most effective emulsifier for a particular emulsion. The type of emulsion formed, O/W or W/O, is also strongly dependent on the choice of emulsifier. For example, a 50/50 mixture of oil and water may form an O/W emulsion or a W/O emulsion depending upon the emulsifier.

The agitation required to form an emulsion can be supplied by different equipment, the simplest being a propeller used to stir the mixture. Other equipment includes homogenizers and colloid mills. In these machines, the shearing forces needed to form the emulsion are obtained by forcing the mixture through a small orifice at high pressure (homogenizer), or by passing the mixture between a close fitting stator and revolving rotor (colloid mill). An emulsion can also be formed by subjecting the mixture to intense ultrasonic irradiation.

In general terms, the stability of an emulsion is a measure of the constancy of its physical properties with time. The time scale associated with significant variation in a physical property may range from seconds to years, depending upon the particular formulation. The changes in the structure of the emulsion that affect the stability are complex; however, a few simple changes can be identified [3,4].

At formation the droplets of the disperse phase are uniformly distributed throughout the emulsion. With time, the droplets may drift upward or downward forming an increase in the disperse phase at the top or bottom of the vessel containing the emulsion. This process is known as creaming, and it is the cause of cream separating from unhomogenized milk. The parameters that affect the rate of creaming are made evident by the Stokes relation for the sedimentation rate v of a spherical particle in a viscous liquid [3,4]:

$$v = 2gr^2(\rho_1 - \rho_2)/9\eta_2 . \quad (2)$$

Here, r is the radius of the droplet, ρ_1 the density of the disperse phase, ρ_2 and η_2 the density and viscosity of the continuous phase, and g is the acceleration due to gravity.¹ Note, for $\rho_2 > \rho_1$, which is normally the case

¹The subscripts 1 and 2 will be used throughout to indicate parameters for the disperse phase and the continuous phase, respectively.

for an O/W emulsion, v is negative and the particle moves upward, while for $\rho_1 > \rho_2$, which is normally the case for a W/O emulsion, v is positive and the particle moves downward. From (2), it is clear that the rate of creaming can be decreased by reducing the size (radius r) of the droplets, making the densities of the two phases nearly the same ($\rho_1 \approx \rho_2$), or increasing the viscosity (η_2) of the continuous phase. Creaming can often be reversed by simply stirring the emulsion.

Another change in the structure of the emulsion that affects the stability is the coagulation of the droplets. Coagulation occurs in stages: first, the droplets form aggregates in a process known as flocculation. Coalescence then occurs with several droplets combining to form a single large drop. This process continues until the two phases form separate layers in the vessel containing the emulsion. At this point, complete demulsification has occurred. The emulsion can then be reformed only by subjecting it to the process (homogenization, ultrasonic irradiation, etc.) used in its initial formation.

Other examples of changes in structure are an inversion, where the emulsion type changes (an O/W emulsion inverts to become a W/O emulsion), and the formation of a multiple emulsion, where both types of emulsions exist simultaneously (O/W and W/O).

The most difficult task in designing an emulsion is usually choosing the ingredients, particularly an emulsifier, and the method of agitation so as to obtain an emulsion with the desired stability.

III. ELECTRICAL PROPERTIES OF EMULSIONS - THEORY

An emulsion can be considered a heterogeneous system consisting of two components, e.g., water and oil. Since the droplets which form the disperse phase of the emulsion are small on the macroscopic scale, their diameters are usually in the range 0.1 to 100 μ , the macroscopic electrical properties of the emulsion can be described by an average permittivity $\bar{\epsilon}$ and an average

conductivity $\bar{\sigma}$.² The average permeability of the emulsion is assumed to be $\bar{\mu} = \mu_0$, since the components are generally nonmagnetic.

Numerous approximate formulas have been proposed for determining the permittivity and the conductivity of heterogeneous systems [6]. Several investigators have examined the accuracy of such formulas for use with emulsions. The most prominent of these is T. Hanai, who carried out an extensive investigation of the electrical properties of emulsions in the late 1950's and the early 1960's [7]. Hanai compared the predictions of various formulas with experimental results, and found that, for certain emulsions, the formula of Bruggeman [8] gave the best agreement. Bruggeman's formula is for lossless dielectrics, with the real relative permittivities ϵ_{r1} and ϵ_{r2} representing the particles (disperse phase) and the matrix (continuous phase), respectively. When the particles are spherical and the volume fraction of the disperse phase is ϕ_1 , Bruggeman's formula for the average relative permittivity $\bar{\epsilon}_r$ of the heterogeneous system becomes

$$\left(\frac{\bar{\epsilon}_r - \epsilon_{r1}}{\epsilon_{r2} - \epsilon_{r1}} \right)^3 = (1 - \phi_1)^3 \frac{\bar{\epsilon}_r}{\epsilon_{r2}} . \quad (3)$$

Hanai showed that Bruggeman's formula also applied for lossy dielectrics. After introduction of the complex relative dielectric factor

$$\xi_r = \xi'_r - j\xi''_r \equiv \epsilon_r - j\sigma/\omega\epsilon_0 , \quad (4)$$

Bruggeman's formula for lossy dielectric becomes

$$\left(\frac{\bar{\xi}_r - \xi_{r1}}{\xi_{r2} - \xi_{r1}} \right)^3 = (1 - \phi_1)^3 \frac{\bar{\xi}_r}{\xi_{r2}} . \quad (5)$$

²The overbar will be used to indicate the average of an electrical parameter for the heterogeneous system.

In the literature on emulsions, equation (5) is sometimes referred to as the Hanai-Bruggeman formula.

It is important to keep in mind that this formula (5) is an approximation. The approximate nature of the formula is due to two factors: the simple physical model used to represent the system (emulsion), and the assumptions made in analyzing the model. For example, the physical model does not account for any special phenomena that may exist at the surface of the particles, such as spatially extended charge layers (Debye screening) that are the result of a diffusion mechanism [9]. The justification for the use of this formula with emulsions rests on its agreement with experimental results, such as those to be presented in the next section of this paper.

The new complex variable η ,

$$\eta \equiv \left(\frac{\bar{\epsilon}_r - \epsilon_{r1}}{\epsilon_{r2} - \epsilon_{r1}} \right), \quad (6a)$$

or

$$\bar{\epsilon}_r = \epsilon_{r1} + \eta(\epsilon_{r2} - \epsilon_{r1}), \quad (6b)$$

is now introduced to put (5) in a form that is more suitable for computation:

$$\eta^3 - \left(1 - \frac{\epsilon_{r1}}{\epsilon_{r2}}\right)(1-\phi_1)^3 \eta - \left(\frac{\epsilon_{r1}}{\epsilon_{r2}}\right)(1-\phi_1)^3 = 0. \quad (7)$$

The three roots of this cubic equation are given by the formula

$$\eta_n = c_1 e^{j(n-1)\pi/3} + \frac{1}{3c_1} \left(1 - \frac{\epsilon_{r1}}{\epsilon_{r2}}\right)(1-\phi_1)^3 e^{-j(n-1)\pi/3}, \quad n = 1, 2, 3, \quad (8a)$$

with

$$c_1 \equiv \left[c_2 \left(1 + \left\{ 1 - c_2 \left[\frac{2}{3} \left(\frac{\epsilon_{r2}}{\epsilon_{r1}} - 1 \right) \right]^3 \right\}^{1/2} \right) \right]^{1/3}, \quad (8b)$$

and

$$c_2 \equiv \frac{1}{2} \left(\frac{\epsilon_{r1}}{\epsilon_{r2}} \right) (1-\phi_1)^3. \quad (8c)$$

The roots of (7) are particularly simple for the two cases $\phi_1 = 0$ and $\phi_1 = 1$. When $\phi_1 = 0$, one of the three roots is always $\eta = 1$; and when $\phi_1 = 1$,

all three roots are $\eta = 0$. Figure 1 shows the tracks the three roots make in the complex η plane as ϕ_1 varies from 0 to 1. Results are presented for both O/W and W/O emulsions. For this example, the constituents of the emulsion are chosen to be lossless (ξ_{r1} and ξ_{r2} real) to simplify the results.

When $\bar{\xi}_r$ is to be computed for a general value of ϕ_1 , with ξ_{r1} and ξ_{r2} specified, an iterative procedure is convenient for selecting the correct root of (7). In this procedure, the three roots are first computed for the case $\phi_1 = 0$; the correct root for this case is always $\eta = 1$ ($\bar{\xi}_r = \xi_{r2}$). Next, ϕ_1 is incremented by a small amount; the three roots are again determined, and the root closest to $\eta = 1$ is selected as the new correct root. This procedure is repeated with the root closest to the previous correct value always selected as the new correct root. When the desired value for ϕ_1 is reached, $\bar{\xi}_r$ is determined from the correct root using (6b). Note, for the example in Figure 1, the correct root is always the one on the horizontal axis within the range $0 < \text{Re}(\eta) < 1$.

The point $\phi_1 = 1$, rather than $\phi_1 = 0$, cannot be used as the starting point in the above procedure. The correct root cannot be selected when $\phi_1 = 1$, since all three roots have the same value $\eta = 0$.

Bruggeman's formula (5) will now be used to compute the electrical properties of a simple emulsion composed of mineral oil and an aqueous sodium chloride solution (saline solution). The numerical results from this example illustrate many of the important electrical characteristics of emulsions. The mineral oil in the emulsion is assumed to be a lossless dielectric with a real relative permittivity which is frequency and temperature independent

$$\epsilon_r = \text{constant} , \quad (9a)$$

and a negligible conductivity

$$\sigma = 0 . \quad (9b)$$

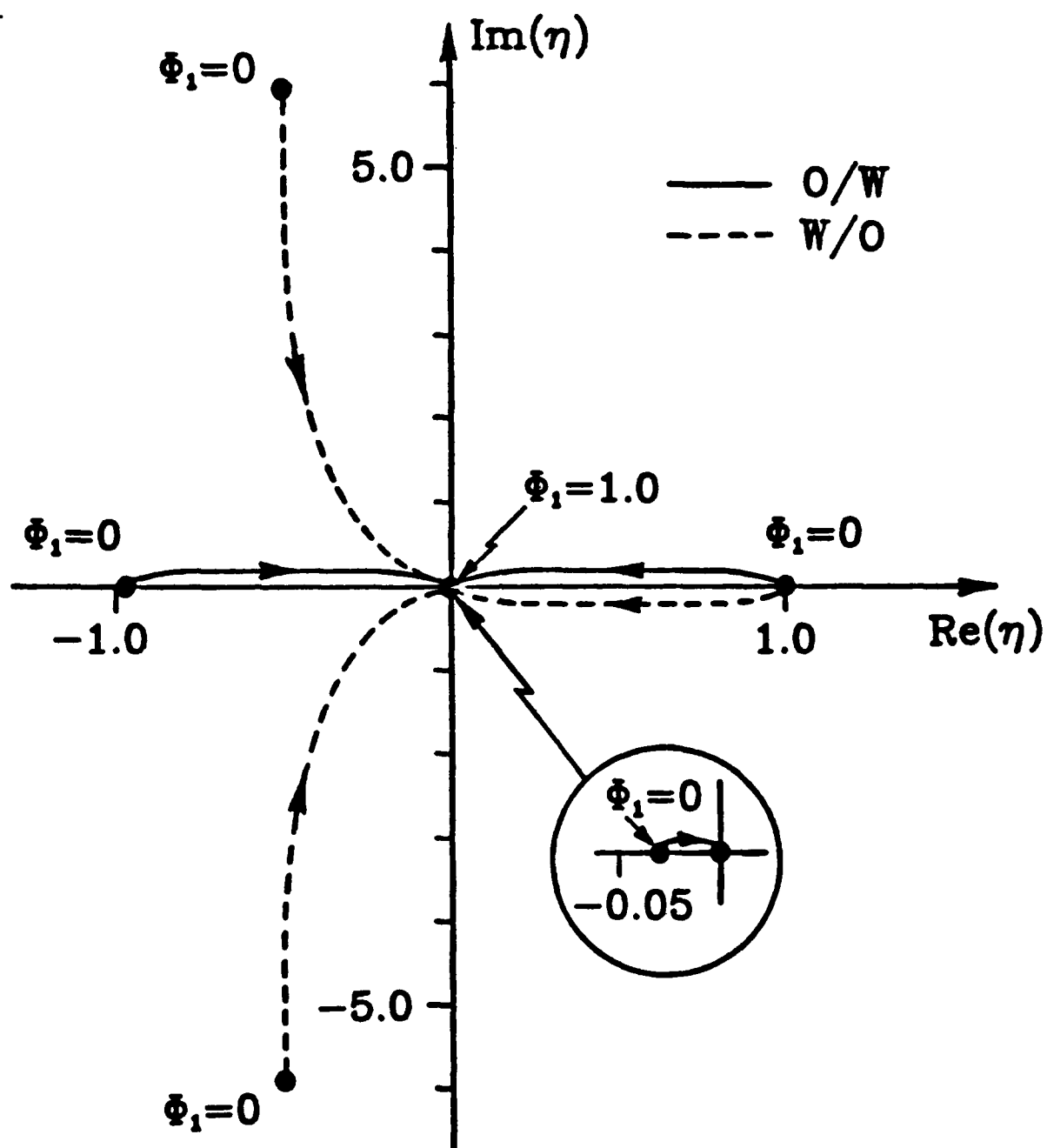


Figure 1. Location of Roots of Cubic Equation for η as a Function of Φ_1 .
 For O/W Emulsion $\xi_{r1} = 2.5$, $\xi_{r2} = 77.0$;
 For W/O Emulsion $\xi_{r1} = 77.0$, $\xi_{r2} = 2.5$.

The saline solution has a complex relative permittivity $\epsilon_r = \epsilon_r' - j\epsilon_r''$ that is well described by the Debye relation for a material with a single relaxation time

$$\epsilon_r = \epsilon_r(T, N) = \epsilon_{r\infty} + [\epsilon_{r0}(T, N) - \epsilon_{r\infty}] / [1 + j\omega\tau(T, N)] , \quad (10a)$$

and a real, frequency independent conductivity

$$\sigma = \sigma(T, N) . \quad (10b)$$

Expressions for the four parameters ϵ_{r0} , $\epsilon_{r\infty}$, τ , and σ , which are functions of the temperature T and the normality N of the saline solution, are given in Appendix A.

The graphs in Figures 2 and 3 show the average relative permittivity $\bar{\epsilon}_r$ and the average conductivity $\bar{\sigma}$ (both are real quantities) as functions of the frequency for two mineral oil, saline solution emulsions. Also shown on the graphs are the factors

$$\bar{\epsilon}_r' = \bar{\epsilon}_r \quad (11)$$

$$\bar{\epsilon}_r'' = \bar{\sigma} / \omega \epsilon_0 .$$

The electrical properties of the emulsions can be described by $\bar{\epsilon}_r$ and $\bar{\sigma}$, or equivalently by $\bar{\epsilon}_r'$ and $\bar{\epsilon}_r''$, and both representations will be useful for the following discussion. Figure 2 is for an oil-in-water (O/W) emulsion with the volume fraction of mineral oil $\phi_1 = 0.5$ and the normality $N = 10^{-3}$ for the saline solution. While Figure 3 is for a water-in-oil (W/O) emulsion with the volume fraction of mineral oil $\phi_1 = 0.5$ and the normality $N = 10^{-3}$ for the saline solution. The relative permittivity for the mineral oil in both emulsions is $\epsilon_r = 2.19$. Note that the ingredients of the two emulsions are the same (50/50 mixture of oil and water), the only difference being that the saline solution forms the continuous phase for Figure 2, while the mineral oil forms the continuous phase for Figure 3. The relative permittivities and conductivities of the two emulsions are seen to be significantly different at all frequencies, with both parameters being generally higher for the O/W emulsion.

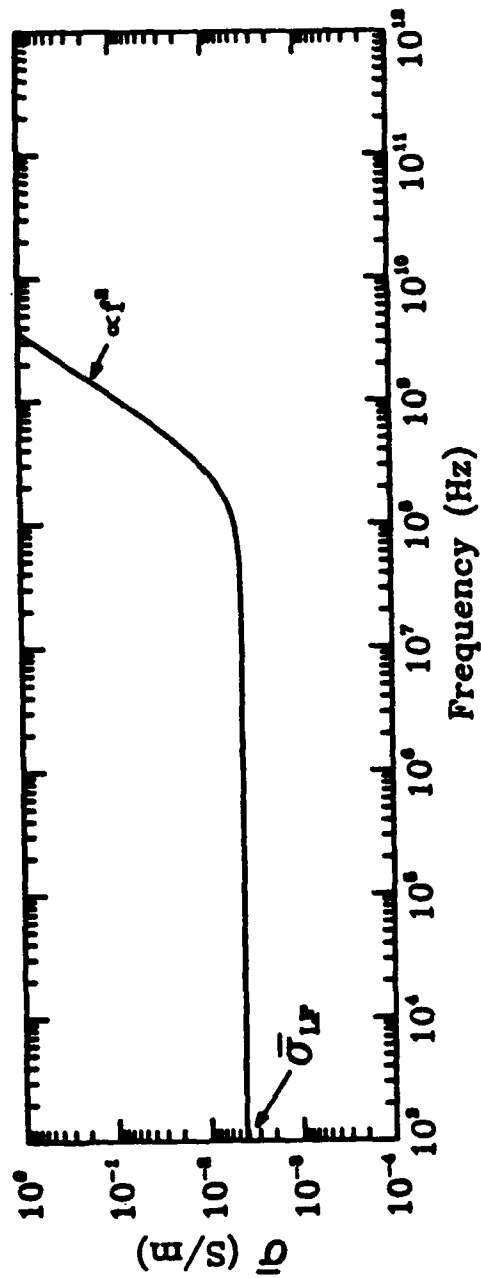
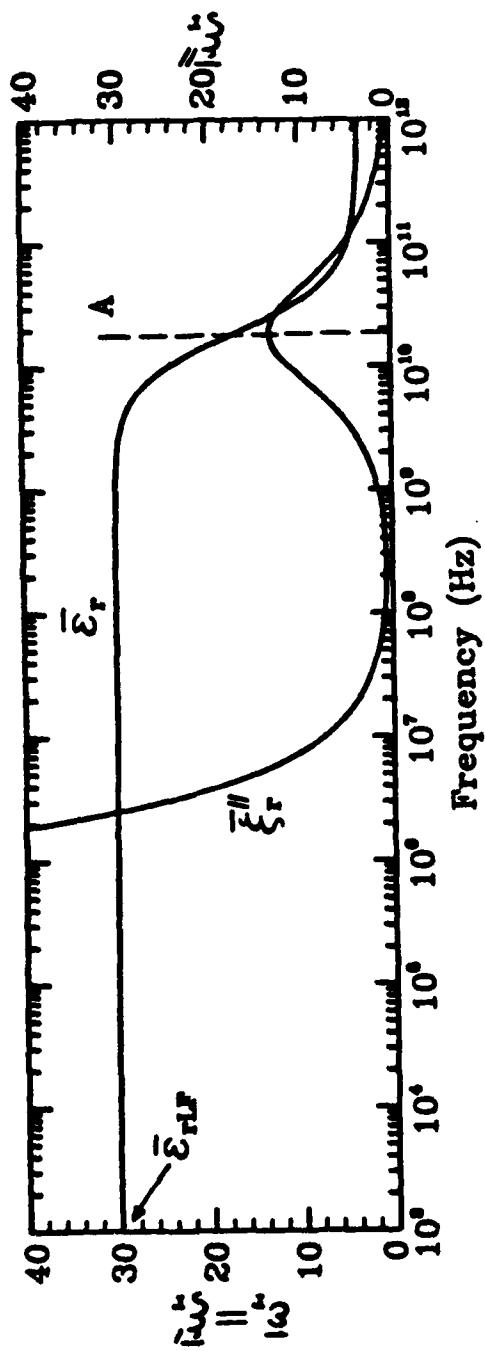


Figure 2. Electrical Constitutive Parameters of a Mineral Oil, Saline Solution O/W Emulsion as a Function of Frequency, $\Phi_1 = 0.5$, $N = 10^{-3}$, $T = 23^\circ\text{C}$.

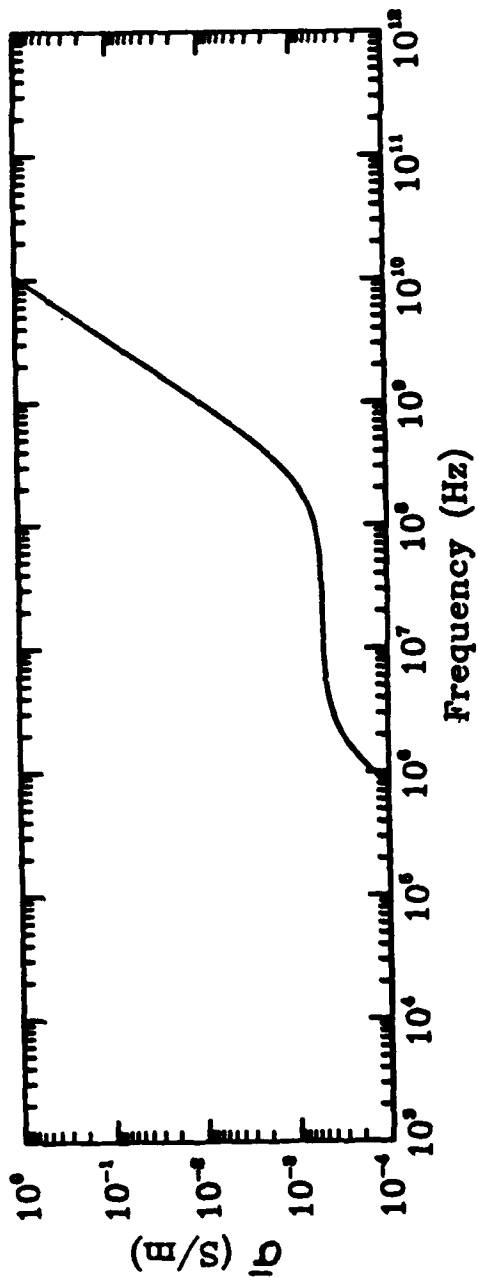
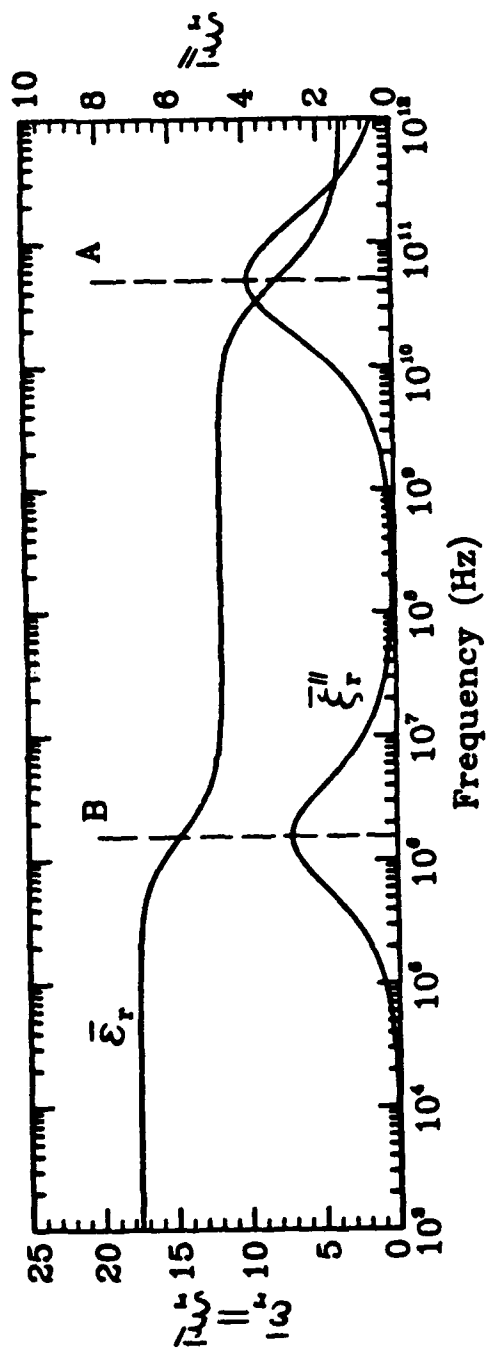


Figure 3. Electrical Constitutive Parameters of a Mineral Oil, Saline Solution W/O Emulsion as a Function of Frequency, $\Phi_1 = 0.5$, $N = 10^{-3}$, $T = 23^\circ\text{C}$.

For both emulsions, the dispersion with frequency in $\bar{\epsilon}'_r$ and $\bar{\epsilon}''_r$ that is centered near $f = 20$ GHz (A on the graphs) is due to the dipolar relaxation of the water molecule. This phenomenon is also the cause of the increase in $\bar{\sigma}$ for frequencies $f \gtrsim 100$ MHz. Additional calculations show that the location and extent of this dispersion are relatively independent of ϕ_1 and N . For the W/O emulsion, Figure 3, there is a second and similar dispersion in $\bar{\epsilon}'_r$ and $\bar{\epsilon}''_r$ centered near $f = 20$ MHz (B on the graph). This dispersion is due to the Maxwell-Wagner effect, which is present for all heterogeneous systems when one or more of the components are conducting [10]. Additional calculations show that the location of this dispersion is a function of ϕ_1 and N . An increase (decrease) in the normality of the saline solution would cause this dispersion to move to higher (lower) frequencies at a rate of approximately one decade for each factor of ten increase in normality. Thus, for normality $N \gtrsim 10^{-2}$, there would be a significant overlap of the two regions of dispersion (A and B in Figure 3) for the W/O emulsion. The dispersion due to the Maxwell-Wagner effect is also present for the O/W emulsion, but it is too small to be seen on the graphs in Figure 2, and in general this dispersion is insignificant for practical O/W emulsions.

Oil-in-Water Emulsions

A material for general use in scale models must have adjustable electrical properties that are dispersionless or have simple, predictable dispersion with frequency. From the above discussion, it is clear that the dispersion for W/O emulsions exhibits a complicated dependence on the constituents of the emulsion (ϕ_1, N). Thus W/O emulsions have limited value as materials for general use in scale models, and they will not be discussed further in this paper. On the other hand, the dispersion in the electrical properties of O/W emulsions is seen to be fairly simple. For frequencies $f \lesssim 3$ GHz, the relative permittivity is approximately constant and equal to the low frequency value $\bar{\epsilon}_{rLF}$,

$$\bar{\epsilon}_r \approx \bar{\epsilon}_{rLF}, \quad (12)$$

while the conductivity is approximately equal to a constant, the low-frequency value $\bar{\sigma}_{LF}$, plus a term which increases as the square of the frequency,

$$\bar{\sigma} = \bar{\sigma}_{LF} + \bar{\epsilon}_{rLF} F_{\sigma} [f(\text{GHz})]^2 . \quad (13)$$

Expressions for the factors $\bar{\epsilon}_{rLF}$, $\bar{\sigma}_{LF}$, and F_{σ} , which appear in these equations, are developed in Appendix B:

$$\bar{\epsilon}_{rLF}(T, N, \phi_1) = \frac{1}{2} \{ 3\epsilon_{r1} + (1-\phi_1)^{3/2} [2\epsilon_{ro}(T, N) - 3\epsilon_{r1}] \} , \quad (14)$$

$$\bar{\sigma}_{LF}(T, N, \phi_1) = \sigma_2(T, N)(1-\phi_1)^{3/2} , \quad (15)$$

$$F_{\sigma}(T, N, \phi_1) = \frac{(2\pi \times 10^9)^2 \epsilon_o [\epsilon_{ro}(T, N) - \epsilon_{r\infty}] \tau(T, N)}{\epsilon_{ro}(T, N)}$$

$$\frac{[2\epsilon_{ro}(T, N) + \epsilon_{r1}][\bar{\epsilon}_{rLF}(T, N, \phi_1) - \epsilon_{r1}]}{[2\bar{\epsilon}_{rLF}(T, N, \phi_1) + \epsilon_{r1}][\epsilon_{ro}(T, N) - \epsilon_{r1}]} . \quad (16)$$

In these equations, ϵ_{r1} is the relative permittivity of the oil phase (9a), and ϵ_{ro} , $\epsilon_{r\infty}$, τ , and σ_2 are the parameters for the saline solution (10a,b).

Numerical values for the average electrical constitutive parameters $\bar{\epsilon}_r$ and $\bar{\sigma}$ of a mineral oil, saline solution O/W emulsion are easily computed from specified ϵ_{r1} , ϕ_1 , N , and T , using equations (12)-(16) and the formulas in Appendix A. However, insight into the dependence of these parameters on the volume fraction of oil ϕ_1 and normality of the saline solution N is more readily obtained from the graphical results presented in Figures 4 and 5. Figure 4 shows the low-frequency electrical constitutive parameters $\bar{\epsilon}_{rLF}$ and $\bar{\sigma}_{LF}$ as functions of ϕ_1 and N at the temperature $T = 23^\circ\text{C}$ (room

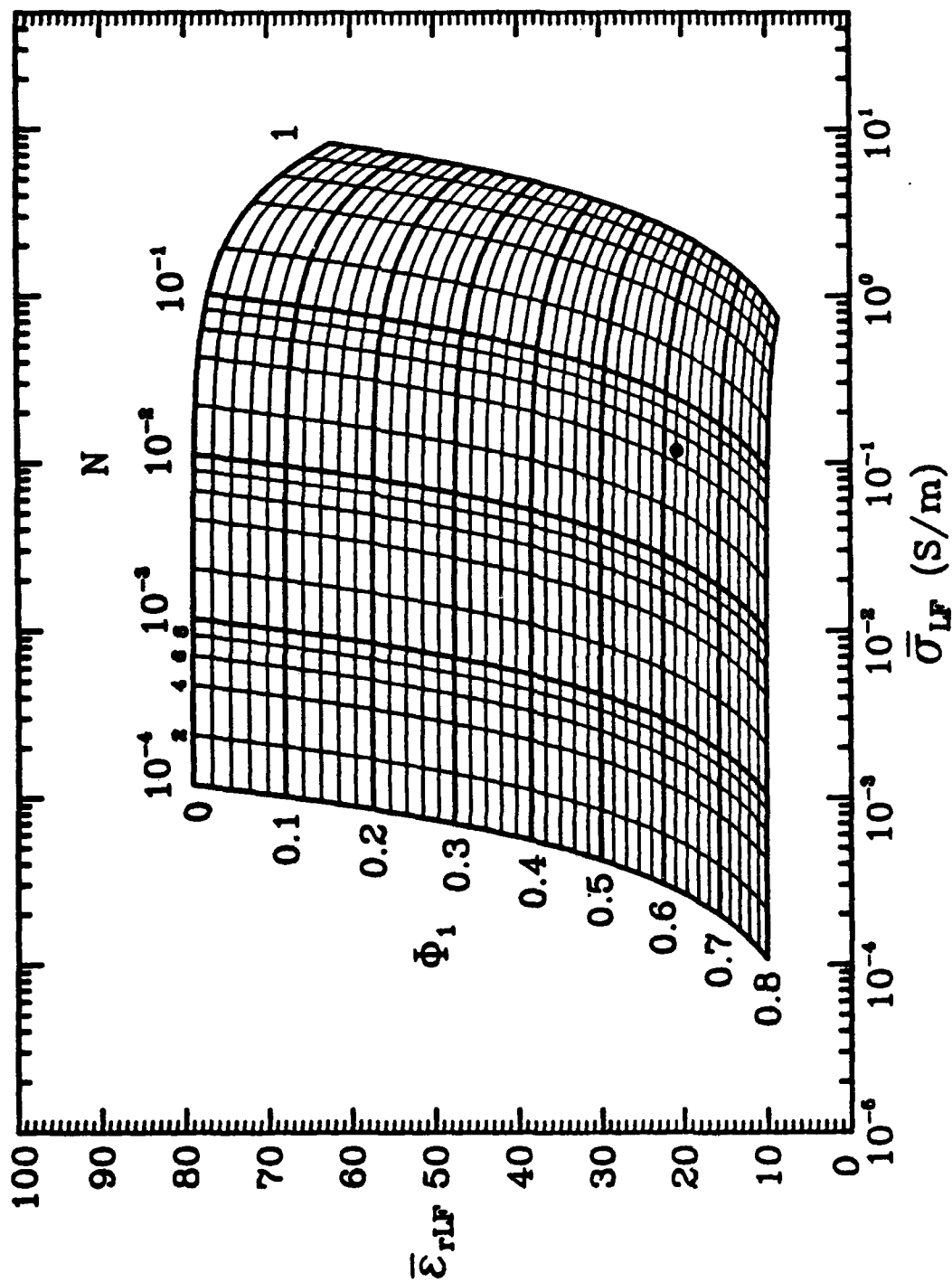


Figure 4. Low-Frequency Electrical Constitutive Parameters of Mineral Oil, Saline Solution O/W Emulsions, $T = 23^\circ\text{C}$.

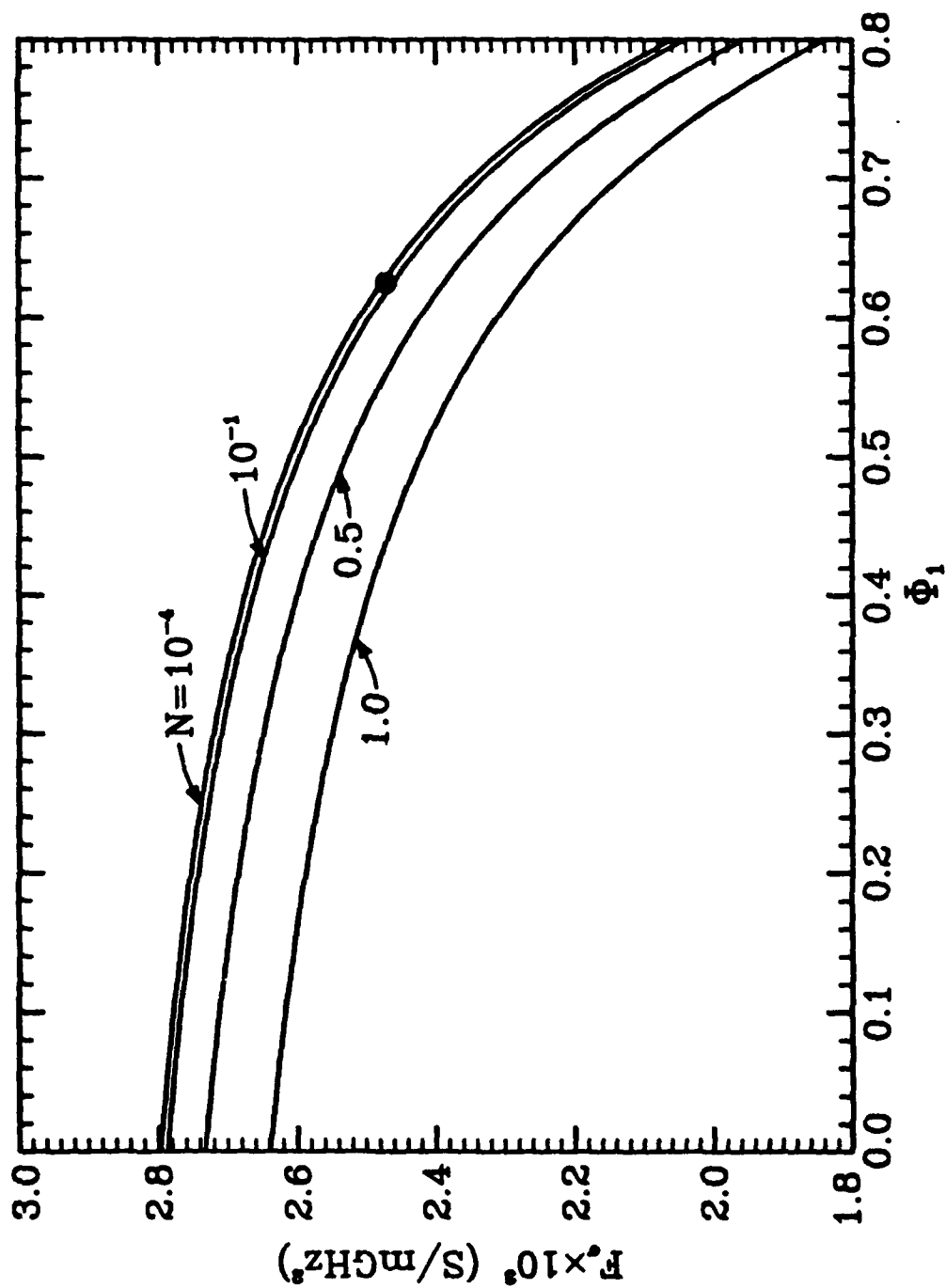


Figure 5. The Coefficient F_e for Mineral Oil , Saline Solution O/W Emulsion, $T = 23^\circ\text{C}$.

temperature). The relative permittivity $\bar{\epsilon}_{rLF}$ is seen to depend mainly on the volume fraction of oil ϕ_1 , while the conductivity $\bar{\sigma}_{LF}$ depends mainly on the normality of the saline solution N . Both parameters are adjustable over wide ranges: $10 \lesssim \bar{\epsilon}_{rLF} \lesssim 80$ and $4 \times 10^{-4} \text{ S/m} \lesssim \bar{\sigma}_{LF} \lesssim 4 \text{ S/m}$.

For normalities $N \lesssim 4 \times 10^{-2}$, the permittivity is nearly independent of the normality, i.e., the curves $\phi_1 = \text{constant}$ are horizontal straight lines. However, at higher normalities the permittivity decreases with increasing normality. The largest decrease is for the pure saline solution, the uppermost curve $\phi_1 = 0$. This phenomenon is thought to be caused by the polar water molecules forming a "hydration sheath" around each ion. The water molecules in the hydration sheath are not free to orient in an applied electric field; this produces a net decrease in the dipole moment per unit volume and, hence, in the permittivity of the solution [11].

In Figure 5, the coefficient F_σ is graphed as a function of ϕ_1 and N at the temperature $T = 23^\circ\text{C}$. F_σ is seen to be fairly independent of the normality for $N \lesssim 10^{-1}$.

IV. A SERIES OF O/W EMULSIONS FOR USE IN SCALE MODELS

The theoretical results presented in the previous section suggest that simple O/W emulsions can serve as materials with adjustable electrical properties for use in scale models. Of course, the theoretical results say nothing about the practicality of formulating these emulsions; this issue will now be addressed.

For a series of emulsions to be of general value for use in scale models, they should satisfy the following requirements.

- (1) The electrical constitutive parameters of the emulsion $\bar{\epsilon}_r$ and $\bar{\sigma}$ should be predictable from simple formulas, such as equations (12)-(16), for a wide range of the parameters ϕ_1 and N .
- (2) All emulsions in the series should be prepared from the same basic ingredients, e.g., a single oil, such as mineral oil; a single emulsifier or a single blend of emulsifiers, etc.

- (3) The emulsifiers used should be nonionic. Recall that nonionic emulsifiers release few free ions into the aqueous phase of the emulsion, while ionic emulsifiers release many free ions. For the conductivity of the emulsions to be determined primarily by the normality of the saline solution, nonionic emulsifiers must be used.
- (4) The ingredients in the emulsion should be relatively nontoxic, nonflammable, noncorrosive, and inexpensive.
- (5) The emulsions should exhibit reasonable stability; that is, the electrical constitutive parameters $\bar{\epsilon}_r$ and $\bar{\sigma}$ should be reasonably constant over the time required for an electromagnetic measurement.

The most difficult task in formulating the series of emulsions was the selection of the proper emulsifier. This task was complicated by the fact that electrolytes, such as sodium chloride, decrease the stability of emulsions stabilized with certain nonionic emulsifiers [12]. Thus, an emulsifier that produced excellent stability for a low conductivity emulsion (low normality for the saline solution) would often produce unacceptable stability for a high conductivity emulsion (high normality for the saline solution).

All of the emulsions tested were formed from heavy white mineral oil (Fisher Scientific No. 0-122, Sabolt viscosity 335/365, density 0.86 gm/ml) and saline solutions prepared by dissolving sodium chloride (Fisher Scientific No. 5271) in distilled water. Measurements of the mineral oil showed that it had negligible loss and a frequency independent relative permittivity $\epsilon_{r1} = 2.19$ ($0 < f < 2.0$ GHz). The low-frequency conductivity for the distilled water was less than 2.0×10^{-4} S/m.

A 200 ml sample of each emulsion was prepared in the following manner. The emulsifier was mixed with the mineral oil, and then the combination was added to the saline solution, while the latter was being stirred. This is known as the "agent-in-oil-method" of forming an O/W emulsion [3]. The sample was then subjected to ultrasonic irradiation while being stirred. The irradiation was provided by an Ultrasonic Dismembrator (Fisher Scientific Model No. 300). The titanium probe of this device, which was placed directly in the

sample, produces approximately 300 Watts of ultrasonic energy at a frequency of 21 kHz. Each sample was exposed for two 4-minute periods; the temperature of the sample rose by about 6-8°C during each exposure.

After formation, the emulsions were monitored over a period of time to determine their stability. The coagulation of oil droplets was determined with a light microscope, while the onset of creaming was determined by simple visual inspection. The final measure of stability was always a continuous or periodic measurement over time of the electrical constitutive parameters $\bar{\epsilon}_r$ and $\bar{\sigma}$.

About 200 emulsions were made and tested using the procedures described above. Several different emulsifiers and emulsifier blends were tried. The selection of emulsifiers was guided somewhat by the use of the "HLB System" (Hydrophile-Lipophile Balance) [3,13-14]. This is a widely used, systematic scheme for narrowing the number of emulsifiers suitable for a particular application. The details of the HLB System are well covered in the references and will not be repeated here.

The most satisfactory of the emulsifiers tested was a blend of the two emulsifiers TWEEN 20 (polyoxethylene sorbitan monolaurate) and SPAN 80 (sorbitan monooleate) with the volume (weight) ratio TWEEN 20/SPAN 80 = 4.5/5.5 (0.926).³ This blend has an "HLB value" of 10.3. Both of these emulsifiers are used in the cosmetics industry and produce no known physical or health hazards [15]. In all of the emulsions to be discussed, the amount of the emulsifier used was ten percent by volume of the mineral oil.

Figures 6, 7, and 8 show the measured electrical constitutive parameters for the final series of emulsions. These measurements were all made using a shielded open-circuited coaxial sample cell formed from a section of 50 Ω Laboratory Precision air line (General Radio GR900). The reflection coeffi-

³TWEEN and SPAN are trademarks of ICI Americas, Inc.

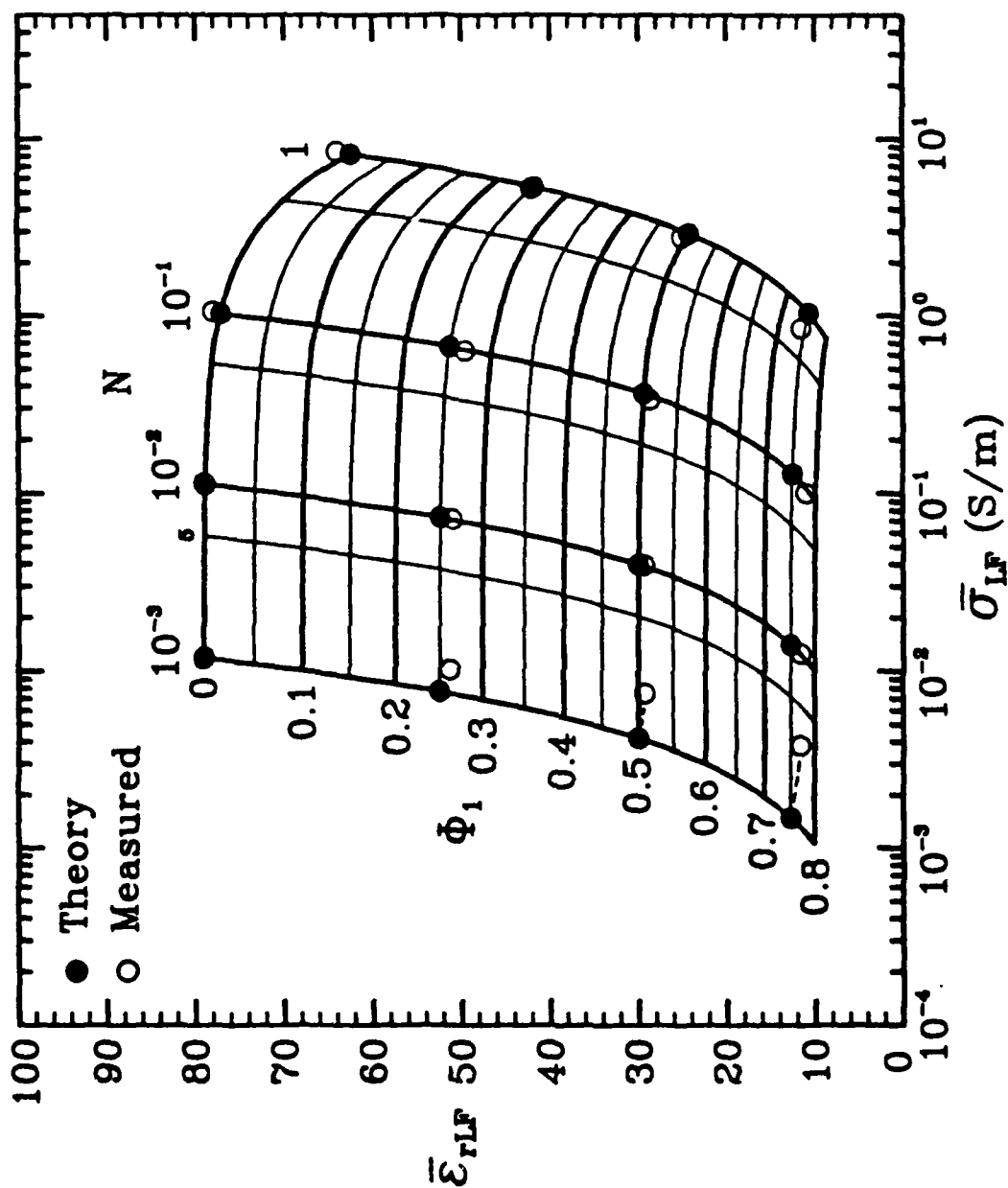


Figure 6. Comparison of Theoretical and Measured Low-Frequency Electrical Constitutive Parameters of Mineral Oil, Saline Solution O/W Emulsions, $T = 23 \pm 1^\circ\text{C}$.

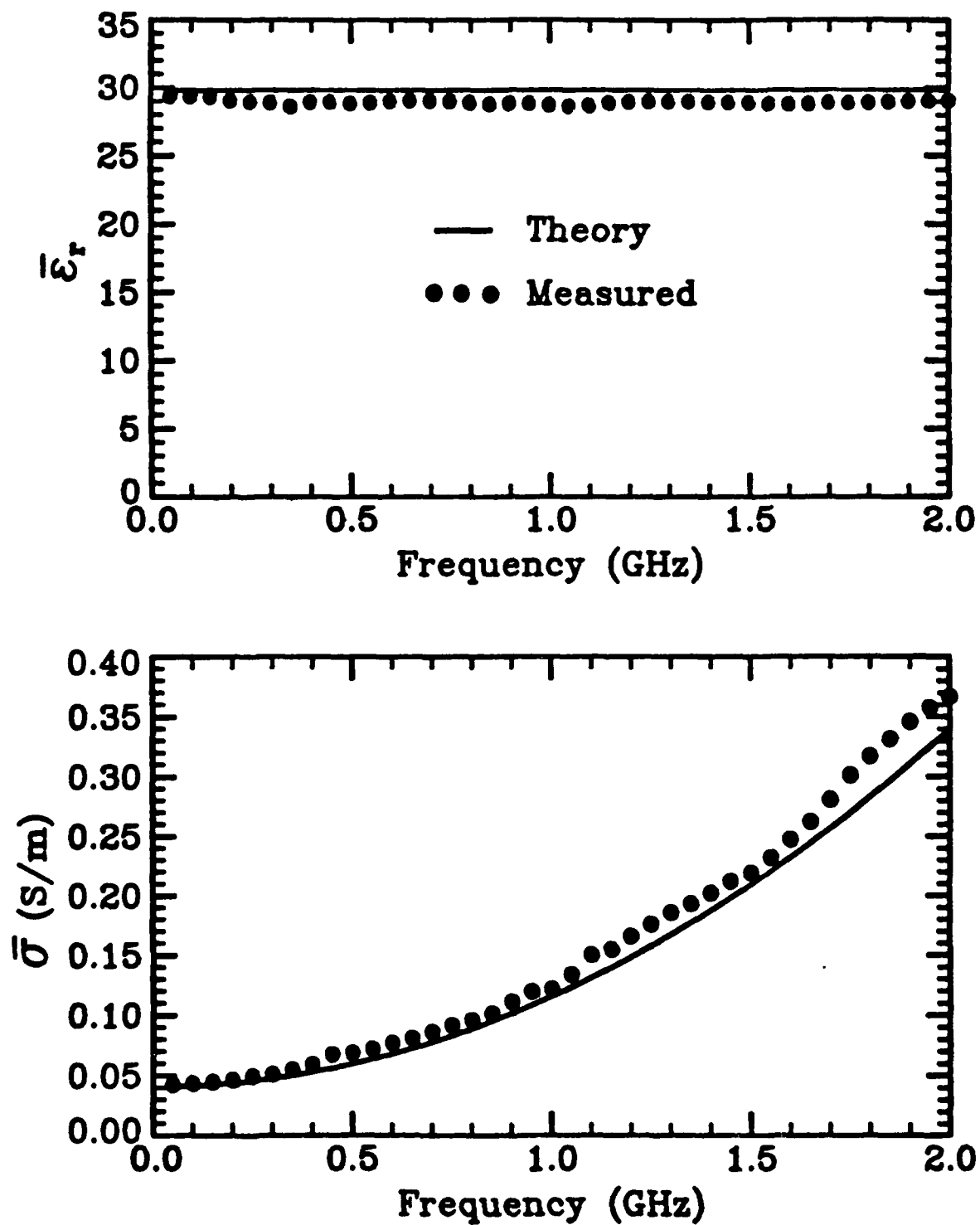


Figure 7. Comparison of Theoretical and Measured Electrical Constitutive Parameters for a Mineral Oil, Saline Solution O/W Emulsion, $\Phi_1 = 0.50$, $N = 10^{-2}$, $T \approx 23^\circ\text{C}$.

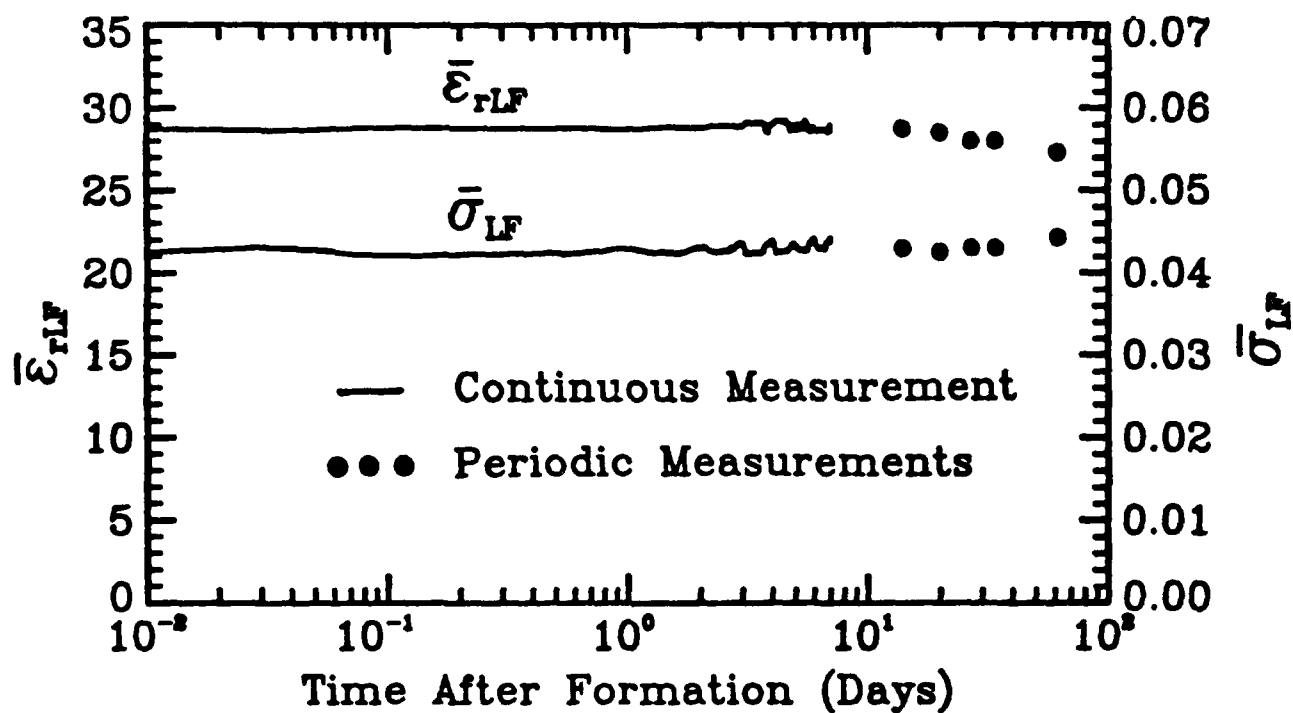


Figure 8. Variation of the Low-Frequency Electrical Constitutive Parameters with Time for a Mineral Oil, Saline Solution O/W Emulsion, $\Phi_1 = 0.50$, $N = 10^{-2}$, $T \approx 23^\circ C$.

cient at the terminals of the cell was measured with a Hewlett Packard model 3577A or model 8409C Automated Network Analyzer. The electrical constitutive parameters of the emulsion were then determined from the reflection coefficient using the procedure described in reference [16].

In Figure 6, the measured low-frequency electrical constitutive parameters $\bar{\epsilon}_{\text{rLF}}$ and $\bar{\sigma}_{\text{LF}}$ are compared with the theoretical values computed from equations (14) and (15). Results are shown for 16 samples: volume fractions of oil $\phi_1 = 0, 0.25, 0.50, \text{ and } 0.75$; and normalities for the saline solution $N = 10^{-3}, 10^{-2}, 10^{-1}, \text{ and } 1.0$. The theoretical values are indicated by the solid dots, while the measured values are indicated by the open dots. The two sets of data are seen to be in good agreement. The only discrepancy is at two points in the lower left-hand corner of the figure: $\phi_1 = 0.5, 0.75; N = 10^{-3}$. For these points, the measured conductivities are higher than the theoretical predictions. This effect is caused by the emulsifier. Even though the emulsifier is nonionic, a small number of free ions, possibly due to impurities, are still released into the aqueous phase of the emulsion. These ions make a noticeable contribution to the conductivity when the emulsifier content is high and the normality of the saline solution is low. This mechanism was verified by additional measurements which showed that a reduction in the amount of emulsifier decreased the discrepancy for the two points in Figure 6.

In Figure 7, the measured constitutive parameters $\bar{\epsilon}_{\text{r}}$ and $\bar{\sigma}$ are graphed as functions of the frequency ($50 \text{ MHz} < f < 2 \text{ GHz}$) for an emulsion with $\phi_1 = 0.50, N = 10^{-2}$. The dispersion in the conductivity with frequency is seen to be well predicted by the theoretical results, equation (13): the conductivity is equal to the low-frequency value plus a term which increases as the square of the frequency.

Figure 8 shows the variation in the constitutive parameters $\bar{\epsilon}_{\text{rLF}}$ and $\bar{\sigma}_{\text{LF}}$ with time after formation of the emulsion. These results are for an emulsion with $\phi_1 = 0.5, N = 10^{-2}$; however, they are typical of the results obtained for other emulsions in the series. The constitutive parameters were measured continuously during the first week after formation (solid line in Figure 8). During this period, the constitutive parameters remained constant except for a

small diurnal variation, which was possibly due to changes in room temperature, or changes in the line voltage to the electronic instrumentation. After the first week, the constitutive parameters were measured periodically at about one week intervals (dots in Figure 8). The emulsion was stirred lightly before each of these measurements. The last measurements were made 60 days after formation; at that time, the electrical parameters showed a small deviation from their initial values.

The constituents of the final series of emulsions are summarized in Table I. A review of the experimental results shows that these emulsions satisfy all of the requirements (1-5), as stated at the beginning of this section, for a series of emulsions to be of general value for use in scale models. The theoretical graphs in Figures 4 and 5 apply to these emulsion, and they can be used to determine the parameters ϕ_1 and N for an emulsion to be used in a particular model.

V. AN EXAMPLE - A MODEL FOR RED-CLAY EARTH

The results of the previous section will now be used to formulate an emulsion for use in an actual scale model. The purpose of this exercise is to illustrate the use of the formulas and graphs presented in the last section and to show how large volumes of emulsions can be formed. Recall that 200 ml volumes of emulsion were used in the previous tests; for the model to be described, several hundred liters of emulsion are required.

The scale model is to be used in experimental tests of ground penetrating radars. A tank filled with the emulsion simulates the earth. Objects, such as pipes, are suspended in the emulsion. The antennas of the radar are placed above the air/emulsion interface, and signals from the radar are used to locate the objects in the emulsion.

A typical Georgia red clay, with a density of 2.04 gm/ml and a water content of 20% by dry weight, was chosen to represent the earth in the full-sized system. The measured electrical constitutive parameters ϵ_r and σ of the clay for frequencies $50 \text{ MHz} < f < 500 \text{ MHz}$ are graphed in Figure 9 (dots).

Table I
Constituents of Oil-in-Water Emulsions
For Use in Scale Models

Oil	Heavy White Mineral Oil Saybolt Viscosity 335/365, Volume Fraction Φ_1.
Saline Solution	Distilled Water with NaCl Added to Adjust Normality N.
Emulsifier	Blend of two Emulsifiers TWEEN 20 and SPAN 80 with the Volume Ratio TWEEN 20/SPAN 80 = 4.5/5.5. Amount of Emulsifier is 10% by Volume of Amount of Oil.

Note that the upper and right-hand scales on the graphs apply to the full-sized system. The relative permittivity of the clay is seen to be fairly constant, while the conductivity increases with frequency.

The scale model is to be one-third size; that is, the scale factor for length in equation (1a) is $k_l = 0.33...$. Therefore, the frequencies in the full-sized system, $50 \text{ MHz} < f_f < 500 \text{ MHz}$, must be increased by the factor (1b) $k_\omega = 3.0$ in the model, $150 \text{ MHz} < f_m < 1.5 \text{ GHz}$. According to equations (1c) and (1e), the relative permittivity in the model should be the same as that in the full-sized system, while the conductivity in the model should be three times that in the full-sized system.

From Figure 9, the relative permittivity of the clay is about 20.5; $\bar{\epsilon}_{rLF}$ for the model should have the same value. Now using Figure 4 or equation (14), and assuming that the normality is low enough that $\bar{\epsilon}_{rLF}$ is practically independent of N , the volume fraction of oil is found to be $\phi_1 = 0.625$.

The conductivity of the clay, Figure 9, increases with the frequency as does the conductivity of the emulsion, equation (13). Thus, there is the possibility that the dispersion with frequency for the clay can be matched with the emulsion. Using Figure 5 or equation (16), and again assuming that the normality is low ($N < 10^{-1}$), the factor F_σ for $\phi_1 = 0.625$ is found to be $F_\sigma = 2.48 \times 10^{-3} \text{ S/m GHz}^2$. From (13), the conductivity of the emulsion is then

$$\begin{aligned}\bar{\sigma} &= \bar{\sigma}_{LF} + \bar{\epsilon}_{rLF} F_\sigma [f_m (\text{GHz})]^2 \\ &= \bar{\sigma}_{LF} + 5.08 \times 10^{-2} [f_m (\text{GHz})]^2 .\end{aligned}\tag{17}$$

Now at the middle frequency $f_f = 300 \text{ MHz}$, the conductivity in the full-sized system is $\sigma = 5.3 \times 10^{-2} \text{ S/m}$; thus at the frequency $f_m = 900 \text{ MHz}$, the conductivity in the model should be $\bar{\sigma} = 1.59 \times 10^{-1} \text{ S/m}$. After substituting this value into (17), $\bar{\sigma}_{LF}$ is found to be

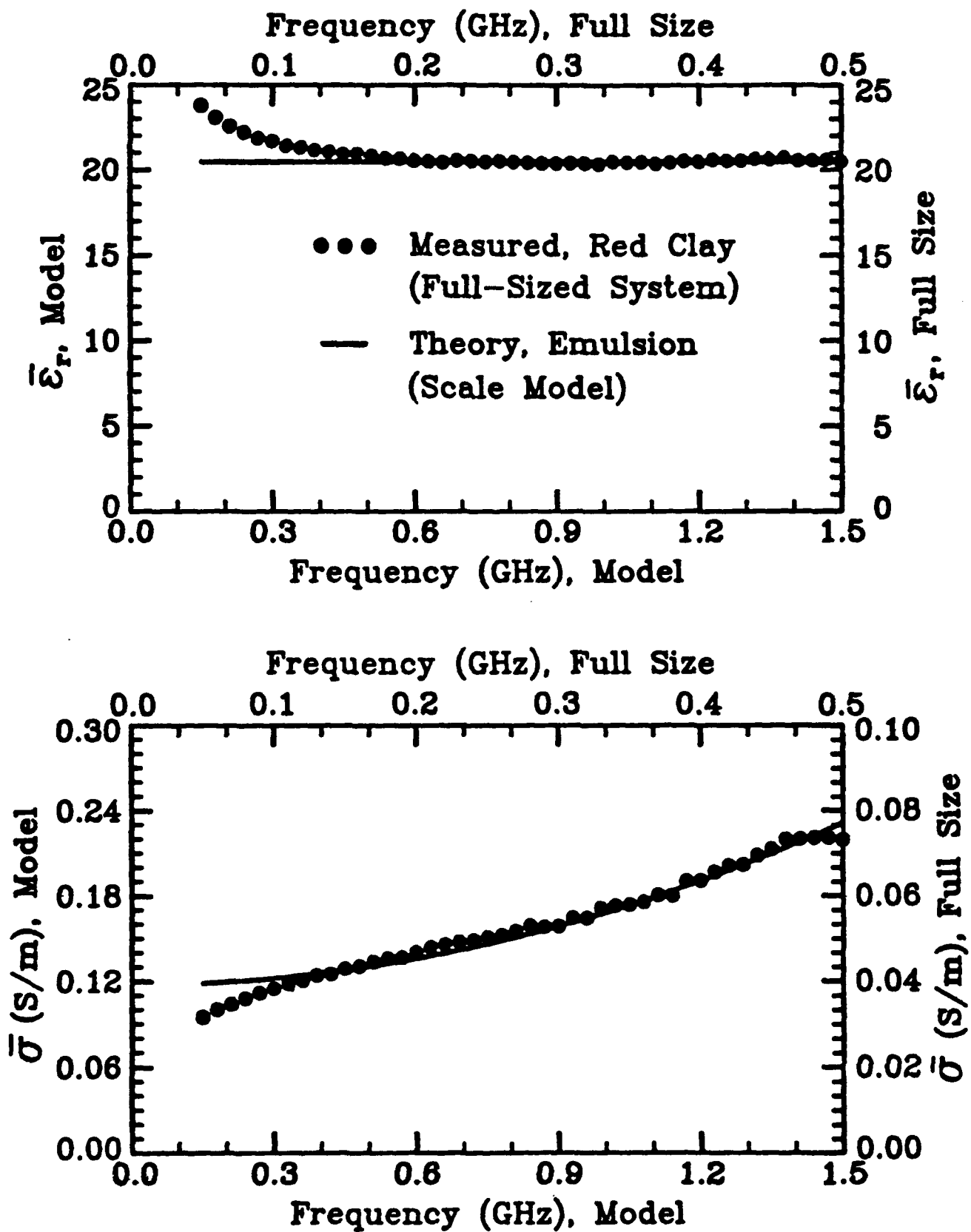


Figure 9. Measured Constitutive Parameters for Red-Clay Earth (Full-Sized System) and Theoretical Constitutive Parameters for Emulsion (Scale Model), $T \approx 23^\circ\text{C}$.

$$\bar{\sigma}_{LF} = 1.18 \times 10^{-1} \text{ S/m} ,$$

and the required conductivity and normality for the saline solution, determined from Figure 4 or equation (15), are $\sigma_2 = 5.13 \times 10^{-1} \text{ S/m}$ and $N = 0.048$.

The parameters for the emulsion to be used in the scale model are summarized below, and the corresponding points in Figures 4 and 5 are indicated by dots:

$$\phi_1 = 0.625 , \quad N = 0.048$$

$$\bar{\epsilon}_{rLF} = 20.5 , \quad \bar{\sigma}_{LF} = 1.18 \times 10^{-1} \text{ S/m} , \quad F_\sigma = 2.48 \times 10^{-3} \text{ S/m GHz}^2 .$$

The electrical constitutive parameters $\bar{\epsilon}_r$ and $\bar{\sigma}$ for this emulsion, computed from equations (12) and (13), are graphed in Figure 9 (solid line). Note that the lower and left-hand scales on the graphs apply to the scale model. The theoretical constitutive parameters for the emulsions are seen to be a good match to the measured constitutive parameters for the red clay over the entire ten-to-one frequency range, when the proper scaling is taken into account.

The tank which contains the emulsion in the scale model has a capacity of 242 liters; thus the ingredients for the emulsion are 142.4 liters of oil; 85.4 liters of saline solution, and 14.2 liters of emulsifier. The mineral oil used in the previous tests was not available in bulk, so an equivalent white mineral oil (Penreco No. 350, Sabolt viscosity 340/360) was substituted. Tap water was also substituted for distilled water in the preparation of the saline solution.

The ultrasonic probe used to form the emulsions in the previous tests is only effective for volumes less than about one liter; thus it could not be used with the 242 liter volume required for the model. Instead, the emulsion was formed by circulating the ingredients through a rotary gear pump (Teel

Model No. 1P776) for twelve hours, at a rate of about 90 liters per minute. The meshing of the gears in the pump provides the shearing action needed to produce the emulsion.

The measured electrical constitutive parameters for the emulsion $\bar{\epsilon}_r$ and $\bar{\sigma}$ are compared with the theoretical results in Figure 10. Data are shown for a small 16 liter trial batch (solid dots) and for the full 242 liter batch used in the model (open dots). Both sets of data are seen to be in good agreement with the theoretical results (solid line). The small difference in the two sets of measured data is thought to be due to the inaccurate measurement of the large volumes of ingredients for the 242 liter batch.

The stability of the emulsion was tested by periodically measuring its electrical constitutive parameters. The graphs in Figure 11 show results measured immediately after formation, one week after formation, and three weeks after formation. After one week, there was negligible change in the parameters. However, after three weeks, the parameters changed significantly. This change is due to creaming of the emulsion, i.e., the oil droplets collecting near the top of the tank. Since the sample used for the measurement was taken from the top of the tank; it had a higher oil content and lower relative permittivity than the initial emulsion. Circulating the emulsion with the gear pump for one hour brought the electrical constitutive parameters back to their initial values.

When the scale model is not in use, the tank is covered to prevent evaporation of the water from the emulsion.

VI. SUMMARY AND DISCUSSION

Emulsions were examined as materials with adjustable electrical constitutive parameters for use in scale models. Both the theoretical and experimental investigations involved only simple emulsions of mineral oil and saline solutions.

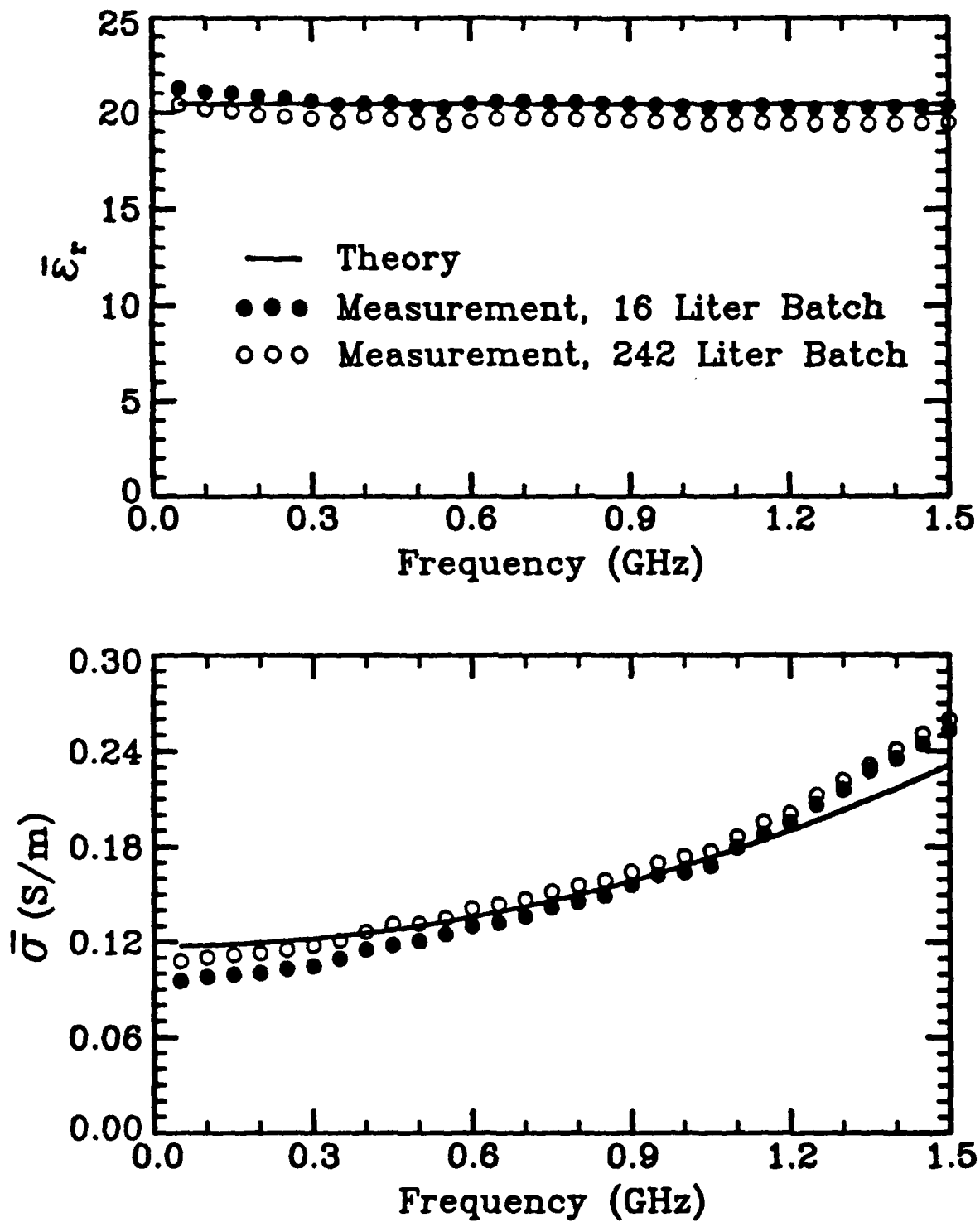


Figure 10. Comparison of Theoretical and Measured Electrical Constitutive Parameter for O/W Emulsion Used to Model Red-Clay Earth, $T \approx 23^\circ\text{C}$.

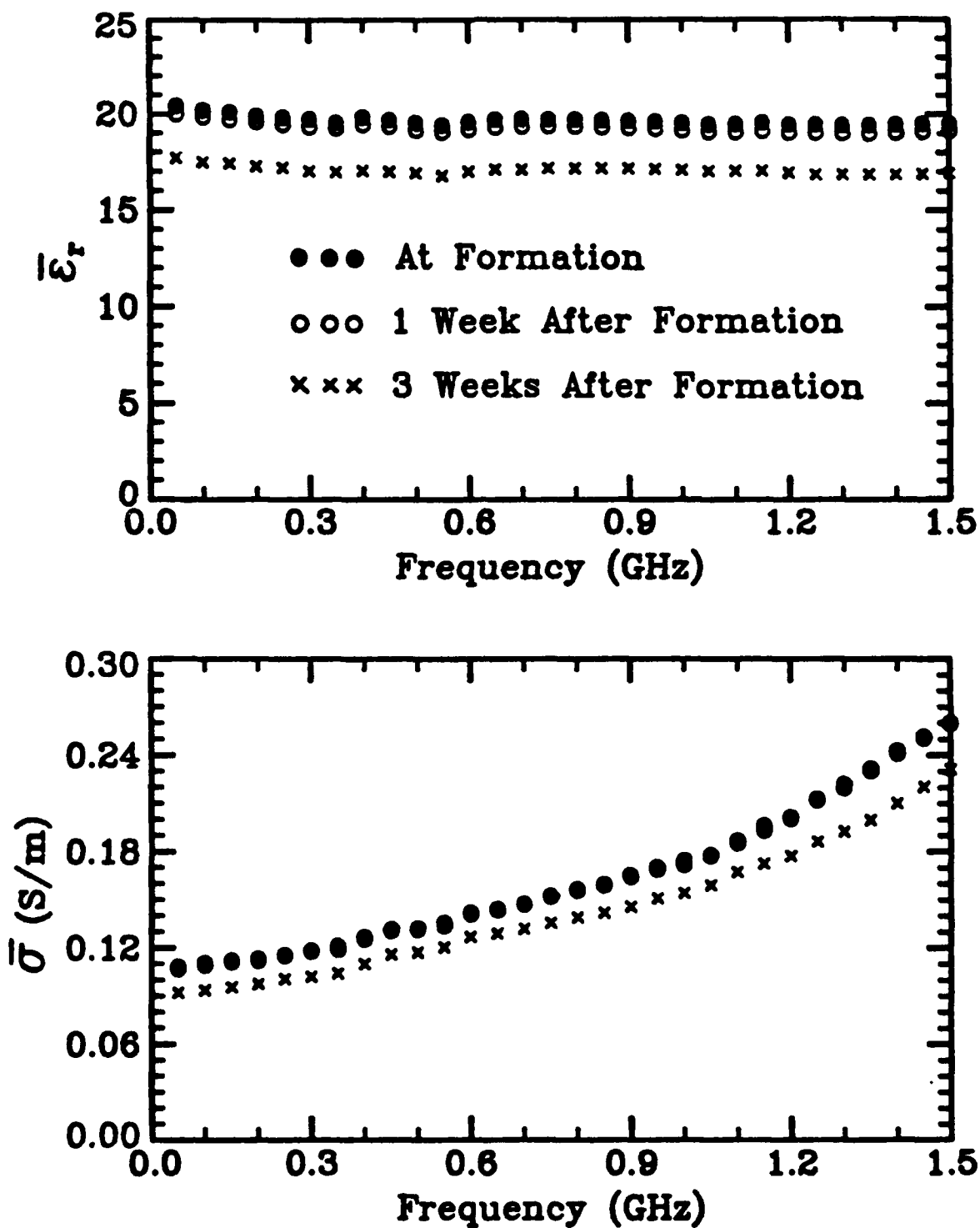


Figure 11. Variation with Time of Electrical Constitutive Parameters of O/W Emulsion Used to Model Red-Clay Earth, $T \approx 23^\circ\text{C}$.

Bruggeman's theoretical formula for the electrical constitutive parameters of a heterogeneous system was applied to the emulsions. For O/W emulsions at frequencies less than about 3 GHz, this formula was shown to reduce to three easily calculable parameters $\bar{\epsilon}_{rLF}$, $\bar{\sigma}_{LF}$, and F_0 . The first two of these parameters are the low-frequency relative permittivity and conductivity, while the third is related to the frequency dependence of the conductivity.

A series of O/W emulsion was formulated for use in scale models; this involved the selection of an appropriate emulsifier. The ingredients for the emulsions are listed in Table I. The electrical constitutive parameters of these emulsions are predicted by the simplified Bruggeman's formula for a wide range of volume fraction of oil ϕ_1 and normality of the saline solution N. The emulsions are stable over a period of one week or more, i.e., the electrical constitutive parameters do not vary significantly during this period.

As an example, an emulsion which is a scale model for red-clay earth was described. This emulsion matches the electrical constitutive parameters of the clay, including the dispersion with frequency in the conductivity, over a ten-to-one frequency range.

The stability of the O/W emulsions can be increased considerably by reducing the rate of creaming, i.e., by reducing the sedimentation rate of the oil droplets. From equation (2), a reduction in the size of the oil droplets is the most effective means of reducing the sedimentation rate (the radius of the droplets is squared in the equation). The size of the oil droplets can be reduced by using a machine specifically designed for preparing emulsions, such as a homogenizer. A homogenizer was not available for use in the experiments reported in this paper. The use of a homogenizer would have the added benefit of reducing the amount of emulsifier required.

The sedimentation rate (2) can also be reduced by making the densities of the two phases nearly equal ($\rho_1 = \rho_2$). This requires an oil which has a density equal to that of water (1.0 gm/ml); silicone based oils are available with this density, but they are very expensive. A third method of reducing

the sedimentation rate and increasing the stability of the O/W emulsion is to increase the viscosity η_2 of the saline solution by the addition of a thickening agent, such as methylcellulose.

The discussion in this paper was restricted to O/W emulsion. W/O emulsions have additional dispersion in their electrical constitutive parameters; this makes them less suitable for scale models which are to be used over a broad band of frequencies. W/O emulsions, however, are suitable for narrow band and single frequency models. W/O emulsions generally can be made with lower relative permittivity and conductivity than similar O/W emulsions, e.g., the emulsions in Figures 2 and 3. W/O emulsions can also be made semisolid or solid by using a heavy grease or wax as the continuous phase.

Emulsion science is an active area of research with vast application in the food, pharmaceutical, cosmetics, etc., industries. The literature on this subject is voluminous. Only the simplest O/W emulsions were examined in this paper; there are certainly other emulsions that have interesting electrical properties and possible electromagnetic applications.

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APPENDIX A

EXPRESSIONS FOR ELECTRICAL PARAMETERS OF AQUEOUS SODIUM CHLORIDE SOLUTIONS

The following expressions for the electrical parameters of aqueous sodium chloride solutions as functions of the normality N and temperature $T(^{\circ}\text{C})$ are from Stogryn [17]:

Static Permittivity

$$\epsilon_{ro}(T, N) = \epsilon_{ro}(T, 0)a(N) \quad (18)$$

$$\epsilon_{ro}(T, 0) = 87.74 - 4.001 \times 10^{-1}T + 9.398 \times 10^{-4}T^2 + 1.410 \times 10^{-6}T^3 \quad (19)$$

$$a(N) = 1.000 - 2.551 \times 10^{-1}N + 5.151 \times 10^{-2}N^2 - 6.889 \times 10^{-3}N^3 \quad (20)$$

Infinite Frequency Permittivity

$$\epsilon_{\infty} = 4.9 \quad (21)$$

Relaxation Time

$$\tau(T, N) = \tau(T, 0)b(N, T) \quad (22)$$

$$\begin{aligned} \tau(T, 0) = & 1.768 \times 10^{-11} - 6.086 \times 10^{-13}T \\ & + 1.104 \times 10^{-14}T^2 - 8.111 \times 10^{-17}T^3 \end{aligned} \quad (23)$$

$$\begin{aligned} b(N, T) = & 1.463 \times 10^{-3}NT + 1.000 - 4.896 \times 10^{-2}N \\ & - 2.967 \times 10^{-2}N^2 + 5.644 \times 10^{-3}N^3. \end{aligned} \quad (24)$$

These formulas apply for $0^{\circ}\text{C} < T < 40^{\circ}\text{C}$, $0 < N < 3$.

Conductivity

Stogryn also provides an expression for the conductivity. However, his expression was found not to agree with measurements, particularly for low values of normality.

DeWane and Hamer provide accurate formulas for determining the conductivity of aqueous sodium chloride solutions at the temperature $T = 25^{\circ}\text{C}$ and data in tabular form for conductivities at other temperatures [18]. The temperature dependence of these data was fit with a linear equation, and the following expression obtained which extends the formulas for $T = 25^{\circ}\text{C}$ to the temperature range $15^{\circ}\text{C} < T < 25^{\circ}\text{C}$:

$$\sigma(T, N) = \sigma(25^{\circ}\text{C}, N)[1 + c(N)(T-25)] \quad (25)$$

$$0 < N < 0.01$$

$$\sigma(25^{\circ}\text{C}, N) = N[12.653 - 8.974 N^{1/2} + 4.682 N \ln(N) + 18.234 N] \quad (26)$$

$$c(N) = 0.0208 \quad (27)$$

$$0.01 < N < 0.1$$

$$\begin{aligned} \sigma(25^{\circ}\text{C}, N) = N(12.653 - 8.974 N^{1/2} + 6.8046 N + 33.567 N^{3/2} \\ - 1.4148 \times 10^2 N^2 + 1.6649 \times 10^2 N^{5/2}) \end{aligned} \quad (28)$$

$$c(N) = 0.0202 \quad (29)$$

$$0.1 < N < 1.0$$

$$\sigma(25^{\circ}\text{C}, N) = N(12.653 - 8.974 N^{1/2} + 11.385 N - 10.403 N^{3/2} + 4.7224 N^2 - 8.2288 \times 10^{-1} N^{5/2}) \quad (30)$$

$$c(N) = 0.0193 . \quad (31)$$

APPENDIX B

APPROXIMATE EXPRESSIONS FOR O/W EMULSIONS

The low-frequency values $\bar{\epsilon}_{rLF}$ and $\bar{\sigma}_{LF}$, (14) and (15), are obtained by examining Bruggeman's formula (5) in the limit $\omega \rightarrow 0$. The parameters in this formula are, from (9a,b)

$$\epsilon_{r1} = \epsilon'_{r1} - j\epsilon''_{r1} \approx \epsilon_{r1}, \quad (32)$$

and from (10a,b) in the limit $\omega \rightarrow 0$

$$\epsilon_{r2} = \epsilon'_{r2} - j\epsilon''_{r2} = \epsilon_{r0} - j\sigma_2/\omega\epsilon_0. \quad (33)$$

After substitution of (32) and (33), (5) becomes

$$\left\{ \frac{(\omega\epsilon_0)^2(\bar{\epsilon}_r - \epsilon_{r1})(\epsilon_{r0} - \epsilon_{r1}) + \bar{\sigma}\sigma_2 + j\omega\epsilon_0[\sigma_2(\bar{\epsilon}_r - \epsilon_{r1}) - \bar{\sigma}(\epsilon_{r0} - \epsilon_{r1})]}{(\omega\epsilon_0)^2(\epsilon_{r0} - \epsilon_{r1})^2 + \sigma_2^2} \right\}^3$$

$$= (1-\phi_1)^3 \left[\frac{(\omega\epsilon_0)^2\bar{\epsilon}_r\epsilon_{r0} + \bar{\sigma}\sigma_2 + j\omega\epsilon_0(\sigma_2\bar{\epsilon}_r - \bar{\sigma}\epsilon_{r0})}{(\omega\epsilon_0)^2\epsilon_{r0}^2 + \sigma_2^2} \right]. \quad (34)$$

Now on equating the real and imaginary parts on both sides of (34) and taking the limit $\omega \rightarrow 0$, the expressions for $\bar{\epsilon}_{rLF}$ and $\bar{\sigma}_{LF}$ are obtained: for the real parts

$$\left(\frac{\bar{\sigma}_{LF}}{\sigma_2} \right)^3 = (1-\phi_1)^3 \frac{\bar{\sigma}_{LF}}{\sigma_2}, \quad (35)$$

or

$$\bar{\sigma}_{LF} = \sigma_2(1-\phi_1)^{3/2}, \quad (36)$$

and for the imaginary parts

$$3 \left(\frac{\bar{\sigma}_{LF}}{\sigma_2} \right)^2 \left[(\bar{\epsilon}_{rLF} - \epsilon_{r1}) - \frac{\bar{\sigma}_{LF}}{\sigma_2} (\epsilon_{ro} - \epsilon_{r1}) \right] = (1 - \phi_1)^3 \left(\bar{\epsilon}_{rLF} - \frac{\bar{\sigma}_{LF}}{\sigma_2} \epsilon_{ro} \right), \quad (37)$$

which on substitution of (36) gives

$$\bar{\epsilon}_{rLF} = \frac{1}{2} \left[3\epsilon_{r1} + (1 - \phi_1)^{3/2} (2\epsilon_{ro} - 3\epsilon_{r1}) \right]. \quad (38)$$

The expression for the coefficient F_o (16) will now be obtained. For frequencies $f \lesssim 3$ GHz and room temperature $\omega\tau \ll 1$, which makes ξ_{r2} approximately

$$\xi_{r2} = \xi_{r2LF} + \Delta\xi_{r2} \approx (\epsilon_{ro} - j\sigma_2/\omega\epsilon_o) - j(\epsilon_{ro} - \epsilon_{r1})\omega\tau. \quad (39)$$

Here $\Delta\xi_{r2}$ represents the deviation of the electrical properties of the saline solution from the low-frequency values. $|\Delta\xi_{r2}|$ is small, typically $|\Delta\xi_{r2}|/\epsilon_{ro} < 0.2$. A similar small deviation $\Delta\bar{\xi}_r$ is anticipated for the electrical properties of the emulsion

$$\bar{\xi}_r = \bar{\xi}_{rLF} + \Delta\bar{\xi}_r. \quad (40)$$

Bruggeman's formula (5) can be written as

$$W(\bar{\xi}_r) = (1 - \phi_1)^3 W(\xi_{r2}), \quad (41)$$

with

$$W(z) \equiv (z - \xi_{r1})^3 / z. \quad (42)$$

For small variations in $\bar{\xi}_r$ and ξ_{r2} , as in (39) and (40),

$$W'(\bar{\epsilon}_{rLF})\Delta\bar{\epsilon}_r = (1-\phi_1)^3 W'(\epsilon_{r2LF})\Delta\epsilon_{r2} , \quad (43)$$

where the prime indicates differentiation with respect to the argument. After performing the differentiation, (43) becomes

$$\Delta\bar{\epsilon}_r = \left[\frac{\bar{\epsilon}_{rLF}(2\epsilon_{r2LF} + \epsilon_{r1})(\bar{\epsilon}_{rLF} - \epsilon_{r1})}{\epsilon_{r2LF}(2\bar{\epsilon}_{rLF} + \epsilon_{r1})(\epsilon_{r2LF} - \epsilon_{r1})} \right] \Delta\bar{\epsilon}_{r2} , \quad (44)$$

which on substitution of (32) and (33) and setting $\Delta\bar{\sigma} = j\omega\epsilon_0\Delta\bar{\epsilon}_r$ becomes

$$\Delta\bar{\sigma} = \left\{ \frac{(\bar{\epsilon}_{rLF} - j\bar{\sigma}_{LF}/\omega\epsilon_0)[(2\epsilon_{ro} + \epsilon_{r1}) - j2\sigma_2/\omega\epsilon_0][(\bar{\epsilon}_{rLF} - \epsilon_{r1}) - j\bar{\sigma}_{LF}/\omega\epsilon_0]}{(\epsilon_{ro} - j\sigma_2/\omega\epsilon_0)[(2\bar{\epsilon}_{rLF} + \epsilon_{r1}) - j2\bar{\sigma}_{LF}/\omega\epsilon_0][(\epsilon_{ro} - \epsilon_{r1}) - j\sigma_2/\omega\epsilon_0]} \right\} \cdot \epsilon_0(\epsilon_{ro} - \epsilon_{rw})\tau\omega^2 . \quad (45)$$

Now it is recognized from (39) that $|\Delta\epsilon_{r2}|$ is only of importance when it is comparable to or greater than $\sigma_2/\omega\epsilon_0$, i.e., when the loss due to the dipolar relaxation of the water molecule is comparable to or greater than the loss due to ionic conduction. Since $|\Delta\epsilon_{r2}| \ll \epsilon_{ro}$, this implies $\sigma_2/\omega\epsilon_0 \ll \epsilon_{ro}$ when $|\Delta\epsilon_{r2}|$ is significant. Thus, in (45) the term $\epsilon_{ro} - \sigma_2/\omega\epsilon_0$ may be replaced by ϵ_{ro} . A similar argument shows that $\bar{\epsilon}_{rLF} - j\bar{\sigma}_{LF}/\omega\epsilon_0$ may be replaced by $\bar{\epsilon}_{rLF}$, making (45)

$$\Delta\bar{\sigma} = \left[\frac{\bar{\epsilon}_{rLF}(2\epsilon_{ro} + \epsilon_{r1})(\bar{\epsilon}_{rLF} - \epsilon_{r1})}{\epsilon_{ro}(2\bar{\epsilon}_{rLF} + \epsilon_{r1})(\epsilon_{ro} - \epsilon_{r1})} \right] \epsilon_0(\epsilon_{ro} - \epsilon_{rw})\tau\omega^2 . \quad (46)$$

When the conductivity of the emulsion is expressed as

$$\bar{\sigma} = \bar{\sigma}_{LF} + \Delta\bar{\sigma} = \bar{\sigma}_{LF} + \bar{\epsilon}_{rLF} F_0 [f(\text{GHz})]^2 , \quad (47)$$

the coefficient F_σ is

$$F_\sigma = \frac{(2\pi \times 10^9)^2 \epsilon_0 (\epsilon_{ro} - \epsilon_{rw}) \tau}{\epsilon_{ro}} \left[\frac{(2\epsilon_{ro} + \epsilon_{rl})(\bar{\epsilon}_{rlF} - \epsilon_{rl})}{(2\bar{\epsilon}_{rlF} + \epsilon_{rl})(\epsilon_{ro} - \epsilon_{rl})} \right]. \quad (48)$$



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